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THE EFFECTS OF ALPHA PARTICLES ON
LOW DENSITY POLYETHYLENE

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Joe A. Swisher

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Department of the Army Project No. 512-10-001
BALLISTIC RESEARCH LABORATORIES

ABERDEEN PROVING GROUND, MARYLAND

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BALLISTIC RESEARCH LABORATORIES

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Terminal Ballistics Laboratory

Department of the Army Project No. 512-10-001

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REPORT NO. 1177

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THE EFFECTS OF ALPHA PARTICLES ON LOW DENSITY POLYETHYLENE

ABSTRACT

✓ Polyethylene samples were exposed to two different size polonium-210 alpha sources for increasing durations of time. The resulting chemical changes were studied as a function of absorbed energy by means of infrared spectroscopy. The radiation induced chemical changes observed in the infrared spectra of the polymer resulted in an increase in absorption at the characteristic frequencies of the new chemical bonds. Two methods were used to correlate the analytical data with the alpha particle exposure. One method involves the characteristic infrared band frequencies and the other, a unique approach, the correlation of the integrated areas of the infrared spectra. The results indicate the potentiality of polyethylene as a detector for alpha particles and a calibration tool for polonium alpha sources in the range of one millicurie to one curie.

INTRODUCTION

This report is the second of a series of studies on the effects of radiation on low density polyethylene. The first report⁽¹⁾ described the dosimetric properties of the polymer when exposed to gamma radiation. This work is a study of the induced chemical changes observed in the infrared spectrum and the resulting dosimetric potential of low density polyethylene when subjected to varying doses of alpha particles from polonium-210 sources. The resulting changes induced by alpha radiation are compared with those obtained by gamma radiation.

During the past fifteen years, several studies on the physical and chemical changes in polymers resulting from exposure to neutrons (pile), electrons, gamma and X-rays have been reported in the literature. In these studies, a few authors have investigated the dosimetric potential of some polymers. However, there is an apparent gap in the information reported. First, a wide variety of physical and chemical methods have been employed to assess the changes in the various types of polymers that have been exposed. Secondly, most studies have been concerned with the above mentioned forms of radiation with little work reported for alpha radiation effects. Thirdly, a paucity of data exists for polymeric materials that will simultaneously detect and measure two different types of radiation by one physical or chemical method.

The structure of low density polyethylene is ill-defined since both amorphous and crystalline forms of molecular arrangement are present in the polymer. The dimethylene group, $(-\text{CH}_2-\text{CH}_2-)$, is the basic repeating monomeric unit, but the existence of ethylenic units, $(-\text{CH}=\text{CH}-)$, and side-chain formation complicates the structure of the polymeric molecule. In high density polyethylene, the crystalline content is large. As a result, the molecules have a more ordered arrangement, less chain branching and are packed tighter than in low density polyethylene. Because of the more ordered arrangement, the radiation effects in high density polyethylene are not identical to that of low density polyethylene.

The effect of methyl through butyl chain branching on the reduction of the crystalline content of polyethylene has been studied by Reding and Lovell⁽²⁾, who prepared copolymers containing ethylene and the necessary alkene to form the

desired branched chain. Their results showed that crystallinity was influenced the least by methyl branches, while the ethyl side chains were the most effective for decreasing the crystalline ratio. All other longer side chains produced effects which were similar, but intermediate between the methyl and ethyl side chains. Their work supported the view of Rugg, et al⁽³⁾, that some of the branches in low density polyethylene are pendant ethyl groups and that the higher crystalline content of high density polyethylene is not a consequence of higher molecular weight, but rather of the lower degree of branching. These workers also determined that the carbon-carbon double bond groups present in low density polyethylene decrease in concentration with increasing molecular weight. It is estimated that the polyethylene used as an alpha detector in this report contained 3.5 pendant methyl groups per 100 carbon atoms. This estimate is based upon studies by Reding and Lovell⁽²⁾ and data supplied by E. I. duPont Company⁽⁴⁾, manufactureres of the polymer film.

Alexander and Toms⁽⁵⁾ have studied the effect of irradiating polyethylene with 2-Mev electrons in the presence of air and have shown that crosslinking of two adjacent polymer chains and main chain scission proceed simultaneously within the polymer chain and are governed by the crystalline content of the polymer. However, when the polymer was irradiated in vacuum or nitrogen atmosphere, only crosslinking occurred. Thus they concluded that crosslinking was independent of oxygen content, while skeletal chain fracture was dependent on oxygen concentration. They postulated that there are two possible methods by which oxygen could produce main chain fracture. One was through the formation of transient peroxides and the other was the formation of the O_2^- radical intermediates, both of which could induce main chain scission.

In polyethylene, there are different forms of unsaturation, which generally may be studied by infrared spectroscopy techniques. The double bonds and their absorption frequencies found in the infrared region are listed in Table I. There is no absorption band found at 705 cm^{-1} for the cis-form of carbon-carbon double bonds, even though there is believed to be equal amounts of both cis and trans-vinylene unsaturation present⁽⁶⁾.

TABLE I
The Types of Double Bonded Carbons and Their Infrared Absorption
Frequencies Found in Polyethylene⁽³⁾.

TYPE OF UNSATURATION	STRUCTURE	WAVENUMBERS
Vinylidene	$RR'C=CH_2$	888 cm^{-1}
Vinyl	$RHC=CH_2$	909 & 993
Trans-Vinylene	$RCH=CHR'$	966
Carbon-Carbon Double Bonds	$C=C$	1644

Pearson⁽⁷⁾ has reported the concentration of the different forms of carbon-carbon double bonds in polyethylene during irradiation. His results showed that hydrogen was evolved continuously during irradiation, and that the degree of crosslinking was a function of the irradiation exposure. However, the trans-vinylene unsaturation concentration increased with irradiation exposure, but did approach a limiting maximum concentration. Thus, he concluded that the trans-vinylene unsaturation was an intermediate in the crosslinking process. Crosslinking, it was concluded, was not influenced significantly by main or side chain scission.

Infrared spectrophotometric determination of the change in double bond concentration during an irradiation experiment is difficult in the 770-1250 cm^{-1} region because of "a continuous increase in the background absorption."⁽⁶⁾ During this study, the above mentioned increase in absorption in this region was found to be proportional to radiation exposure, a favorable relationship for further evaluation. Carbon-carbon double bonds have a stretching frequency found at 1644 cm^{-1} , which may be observed for the total amount of unsaturation present in a polymer as a function of energy. This frequency does not distinguish the different types of unsaturation; only the total unsaturation.

The irradiation of polyethylene in air produces several chemical and physical changes in the polymer. The oxygen reactions contribute to the main and side chain scission, resulting in a variation in the physical strength

characteristics of the polymer. The carbonyl and hydroxyl bonds formed and their growth in irradiated polyethylene is shown vividly in the infrared spectrum determined at 666 to 5000 cm^{-1} . The formation of the carbonyl and hydroxyl bonds can be explained mechanistically by analogy with the work of Luongo⁽⁸⁾ concerning the changes observed in the infrared spectra during the thermally induced oxidation of both high and low density polyethylene. This work suggested that peroxide (OOH) formation, evident at 3555 cm^{-1} , is an intermediate step in the formation of hydroxyl (OH) and carbonyl (C=O) bonds found at approximately 3400 and 1700 cm^{-1} respectively. Both of these absorption bands are very broad, and an enlargement of the carbonyl band reveals it is a composite of many absorption bands at different frequencies. The frequency at which the C=O bond absorbs is governed by the attached molecular structure. Thus a great deal of information may be obtained concerning the oxidized polymer, by using a high resolution spectrophotometer and observing the different carbonyl frequencies in the infrared spectrum. A detailed study of carbonyl frequencies has been reported by Rugg, *et al.*,⁽⁹⁾. The characteristic carbonyl frequencies are summarized in Table II.

TABLE II
Characteristic Carbonyl Frequencies⁽⁹⁾.

TYPE	STRUCTURE	MICRONS	WAVENUMBERS
Anhydrides	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{O}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{R}'$	5.692	1756 cm^{-1}
Esters	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{OR}'$	5.721 5.754	1747 1737
Aldehydes	$\text{R}-\overset{\text{H}}{\underset{\text{O}}{\text{C}}}=\text{O}$	5.772	1732
Ketones	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{R}'$	5.798 5.812	1724 1720
Acids	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{OH}$	5.839	1712

The mechanism proposed by Luongo for the thermal oxidation of both high and low density polyethylene is listed below:

1. $\text{RH} \xrightarrow{\text{Activation}} \text{R}\cdot + \text{H}\cdot$
2. $\text{R}\cdot + \text{O}_2 \longrightarrow \text{ROO}\cdot$
3. $\text{ROO}\cdot + \text{RH} \longrightarrow \text{ROOH} + \text{R}\cdot$
4. $\text{ROOH} \longrightarrow \text{RO}\cdot + \cdot\text{OH}$
5. $\text{ROO}\cdot + \text{HA} \longrightarrow \text{ROOH} + \text{A}\cdot$
6. $\text{R}\cdot + \text{A}\cdot \longrightarrow \text{RA}$

(The dot represents a free or active bond.)

Steps five and six indicate that the reaction may be inhibited by the addition of antioxidants, represented by the symbol HA.

Luongo has reported the presence of peracid and perester carbonyls in heated polyethylene and has found that the concentration of both diminish upon cooling. Thus their absorption coefficients are dependent on temperature, which may indicate that their existence (hence their observation), is dependent on the state of the polymer⁽⁸⁾. Also, it is conceivable that they are intermediates in the accelerated oxidation processes. During the latter stages of oxidation, a carbonyl band appeared at 1685 cm^{-1} which was assigned to an α - β unsaturated ketone group. Thus, he concluded that at these latter stages of oxidation, the oxygen attack is located at an alpha carbon.

The assignments reported by Luongo in the absorption region between 1600 - 1800 cm^{-1} for low density polyethylene are listed in Table III.

Infrared absorption has been observed⁽⁸⁾ at 1415 cm^{-1} and this frequency has been assigned to carbonyl formation between two methylene groups in long aliphatic chains. This is actually a methylene deformation vibration influenced by an adjacent carbonyl group. After the initial oxidation, a broad band of medium intensity was observed between 1125 and 1050 cm^{-1} , and was assigned to the various C-OH, alcoholic components and C-O-C, ether linkages⁽⁸⁾. The band at 3620 cm^{-1} is a combination band from lower frequency C-H vibration⁽¹⁰⁾, and stays relatively constant during irradiation. Thus, it is used as a reference intensity in the report.

TABLE III
Assignments in the 1600-1800 cm^{-1} Absorption Region
for Low Density Polyethylene (8).

TYPE	STRUCTURE	WAVENUMBERS
Peracid Carbonyl	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{OOH}$	1785 cm^{-1}
Perester Carbonyl	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{OOR}'$	1763
Ester Carbonyl	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{OR}'$	1740
Aldehyde Carbonyl	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{H}$	1728
Ketone Carbonyl	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{R}'$	1715
Acid Carbonyl	$\text{R}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{OH}$	1705
α - β Unsaturated Ketone Carbonyl	$\text{R}-\text{CH}=\text{CH}-\overset{\text{O}}{\underset{\text{O}}{\text{C}}}-\text{R}'$	1685
Carbon-Carbon Double Bonds	$\text{C}=\text{C}$	1640

The uniform oxidation of polyethylene by irradiation is dependent upon a supply of oxygen. If the polymer is too thick and the exposure dose rate is too high, the oxygen trapped within the polymer will be consumed at a rate greater than the rate of diffusion of oxygen into the polymer. Thus there will be less oxidation, in comparison to the amount that could occur per unit dose, if stoichiometric amounts of oxygen were available. Therefore, to relate the amount of incident radiation to changes in the absorption of bands in the spectra of polyethylene, one should use a polymer thickness that allows the maximum amount of oxidation to occur per unit dose. Initial survey work on this problem at these Laboratories, using a Cs-137 source having a dose rate of 3.3×10^4 roentgens per hour at 1 cm, indicated that the oxygen replenishment rate was a problem if the thickness of the film was greater than 10 mils.

Broadway⁽¹¹⁾ has reviewed studies in the field of radiation damage to organic compounds, particularly polyethylene, and found most authors agree that resultant effects are dependent on the energy range of the radiation rather than the type of radiation. The "equal energy absorption-equal damage" concept is believed to be true for organic materials where ionization and excitation is produced by the secondary charged particles and photons resulting from interaction of the primary radiation with atoms of the organic materials^(11, 12).

This "equal energy-equal damage" concept may be valid for most physical measurements and some chemical tests; however, it is important to recognize that although the changes in physical and some chemical properties lend credence to this concept, the chemical mechanisms occurring as a result of different types of radiation are not identical. Insight into these chemical mechanisms may be obtained by infrared spectrophotometry.

Shinohara, et. al.⁽¹³⁾, investigated the effect of deuterons and gamma rays on the infrared spectra of polyethylene. The spectra revealed chemical differences in the effect of the two types of radiation. In the sample irradiated with deuterons, trans-vinylene unsaturation (964 cm^{-1}) increased faster than carbonyl formation (1720 cm^{-1}); gamma radiation yielded large carbonyl formation, with less trans-vinylene unsaturation. Their explanation is as follows: "In the samples irradiated by deuterons, the specific ionization along the track is very high compared with that due to the secondary electrons from Co-60 gamma rays. The density of the polymer radicals and free hydrogen atoms will also be very high, and their mutual recombination will occur so frequently, that the formation of unsaturation will be very prominent. In the case of irradiation by gamma rays, the specific ionization is low, and radicals and hydrogen atoms will be distributed some distance apart from each other. Therefore, their chance of mutual recombination will be much less than in the case of deuteron irradiation. The chance of reaction of polymer radicals with oxygen atoms, on the other hand, will be about the same, resulting in the relative increase of carbonyl groups."

In Charlesby's⁽¹⁴⁾ discussion of alpha particles and protons of high energy, he states that the principle effect of these particles is to produce ionization and excitation even though they can cause displacement of the nuclei by collision. Furthermore, the rate of energy loss depends fundamentally on the velocity and charge of the ionizing particle and not on its mass. These particles behave in a manner similar to electrons of the same energy and charge. However, the relatively low velocity of a proton or alpha particle of a few Mev results in intense ionization and short range, and as the proton or alpha particle is slowed down, the ionization density increases rapidly.

The work reported here is concerned with the chemical effects induced in polyethylene by alpha particles. The study was performed to establish a relationship between incident radiation energy and changes in absorbance in the bands of the spectra of low density polyethylene. Using standard samples of polyethylene and known exposures, a relationship was found whereby the amount of exposure of other samples could be determined.

INSTRUMENTATION AND MATERIAL

The two polonium-210 alpha sources used for irradiation were supplied by the Monsanto Chemical Company, Mound Laboratory, Miamisburg, Ohio. The sources were calibrated by the supplier and the initial activities were 0.848 curies and 3.51 millicuries.

Both source containers were 0.25 inches high and made of 304 stainless steel. The polonium-210 in each source was covered with a window of 0.00013-inch stainless steel. The 0.848-curie source had a window diameter of 0.375 inch and a maximum alpha energy of 3.72 Mev. The 3.51-millicurie source had a window diameter of 0.25 inch and a maximum alpha energy of 3.61 Mev. Both sources were leak and vacuum tested to 0.1 mm Hg.

The average energy of the alpha particle emitted from the source and incident upon the polyethylene surface was calculated to be 2.3 Mev. This calculation was performed using the alpha energy spectrum curve and source geometry supplied by Mound Laboratory. The total alpha exposure per polymer

sample was calculated in ergs/cm^2 . This enabled a comparison of the energy effects upon the polymer from the two geometrically different sources. The gamma radiation from the Po-210 sources was determined and found to be negligible at the exposure times used for these polymer studies.

The low density polyethylene was supplied by E. I. duPont de Nemours and Company in film of uniform thickness of $4.3 \text{ mils} \pm 0.2 \text{ mils}$. The average crystalline content of this polyethylene was 50 per cent, and had a density of 0.918 grams/cm^3 (4). The samples were prepared in rectangular form in sizes 1.25×2.0 inches.

A Perkin-Elmer Corporation, model 13, double-beam, ratio recording spectrophotometer, with sodium chloride optics covering the spectral range of 666 to 5000 cm^{-1} , was used for all spectral measurements.

The Perkin-Elmer model 194 printing integrator was attached to the Leeds and Northrup model G recorder, which is a component of the model 13 spectrophotometer. The integrator has a count rate of 0-6000 counts per minute, with 5-digit printout. Linearity was within $\pm 0.3\%$ averaged over full scale. The integrating mechanism is a velocity-servo computing system and is calibrated to the recorder by a strobe-tach indicator.

The sources were positioned over the polymer by a tripod holder. The tripod legs were adjustable in order that the distance from source to polymer could be varied. Figure 1 shows the source and polymer in the irradiation position.

The polymer was mounted over a 0.25-inch hole on 9-mil cardboard. This allowed one to handle the polymer conveniently and served as a reference marker for positioning the source and polymers. Only that part of the polymer located directly beneath the alpha source was utilized for the infrared measurement and correlation studies.

All irradiations were performed in the presence of air, at atmospheric pressure and at a temperature of $27^\circ \pm 5^\circ$ centigrade.

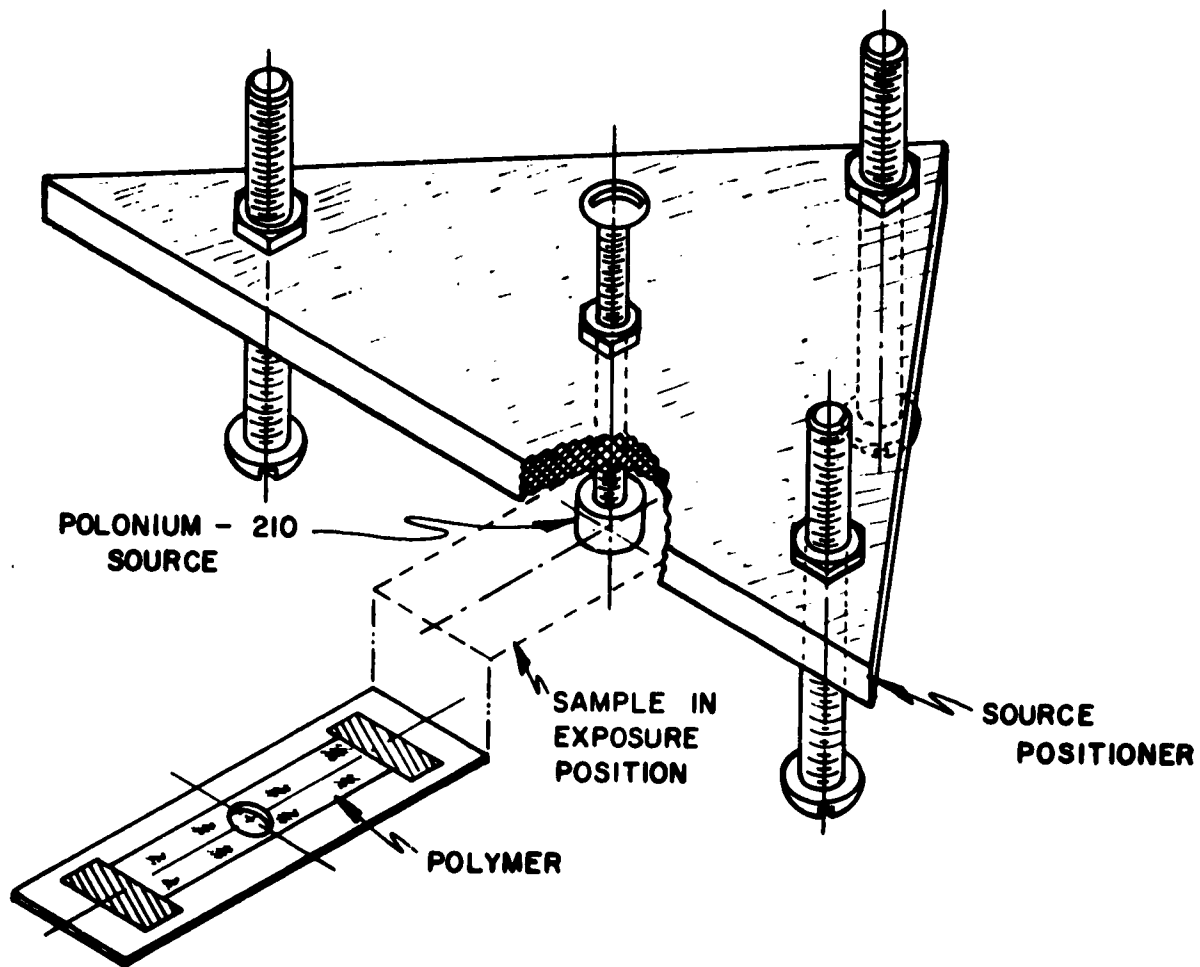


FIGURE 1. POLONIUM - 210 SOURCE , POSITIONER
AND MOUNTED POLYMER SAMPLE

EXPERIMENTAL METHOD AND RESULTS

Prior to irradiation with alpha particles, the specimens were mounted on thin cardboard holders. Next, an infrared standard spectrum between $666\text{-}5000\text{ cm}^{-1}$ was obtained on each sample prior to exposure. When the standard spectrum of the unirradiated polyethylene was determined, the area above the spectral trace was obtained by integration between $666\text{-}5000\text{ cm}^{-1}$. These runs were performed in order to have a standard for evaluation. After standardization, the samples were exposed to the alpha particles. For the spectral analysis, the mounted polymer was attached to brass plates which had a 0.25-inch diameter hole that was aligned in the infrared beam so as to be centered on the monochromator slits. These plates acted as a beam attenuator and a mounting device so that only the area which was exposed would be analyzed.

Each exposure was conducted for a specific length of time and the exposed energy determined. The samples were then analyzed by infrared spectroscopy and the area above the spectral trace was again integrated.

Various absorption changes occur in the infrared spectrum when low density polyethylene is irradiated with alpha particles, and these changes are located in basically three regions of the spectrum that are summarized in Table IV.

TABLE IV
The Three Basic Wavelength Regions Where Absorption Occurs in
Alpha Irradiated Low Density Polyethylene.

WAVELENGTH REGION	ABSORPTION BANDS	REF.
3000 to 3600 cm^{-1}	OOH and OH linkages	8
1600 to 1800 cm^{-1}	various C=O linkages and unsaturation, -C=C-	8
800 to 1400 cm^{-1}	(1) different types of unsaturation, and/or	3
	(2) "a continuous increase in background absorption," and/or	6
	(3) this general depression "can be assigned to ether groups and alcoholic C-OH linkages which are among the oxidation products."	10

Figure 2 shows a composite of the infrared spectra of the unirradiated and alpha exposed polyethylene. The significant differences in the spectra, as compared with the standard are marked with their respective absorption bonds. From this composite, it is apparent that two potential techniques exist for using low density polyethylene as a calibration tool for alpha particles: first, correlation of the increase of carbonyl formation ($C=O$) at one frequency (1720 cm^{-1}) in terms of transmittance vs. dose; and secondly correlation of the overall change in the area of the spectrum between two wavenumbers (666 to 5000 cm^{-1}) as a function of dose.

The former method is useful and the technique is uncomplicated. The analysis is based upon the change in absorption of the carbonyl band. This frequency region is at 1720 cm^{-1} and is primarily attributed to the ketone carbonyl. The densitometer unit attached to the infrared spectrophotometer was used and the results are shown in Figure 3.

The latter method is useful for both large and small exposures, and results in a tabulation which can be readily correlated to exposed dose. Thus, one may integrate the change in area either above or below the spectral trace using either the 100 or 0% transmittance line as the base line, and plot the increase or decrease in integrator units (area) as a function of absorbed energy. Integration was performed over the region from 666 to 5000 cm^{-1} and from the spectral trace to the 100% transmittance line. Any subsequent changes in the polyethylene spectra, as a result of exposure energy, would yield a positive number. Figure 4 shows the results of plotting integrator units vs. exposure energy between the above mentioned wavenumbers.

Each method does have certain advantages which the other does not possess. In the first method, the main advantages are speed, simplicity and less instrumentation required, while it suffers from the inherent disadvantage that it is less sensitive for low doses. The second method has the advantage of increased sensitivity. This advantage is the result of utilizing not only the change in the carbonyl band, but all of the spectral changes induced in low density polyethylene by the alpha particles. These reactions are listed in Table IV. However, the method does involve the incorporation of one extra

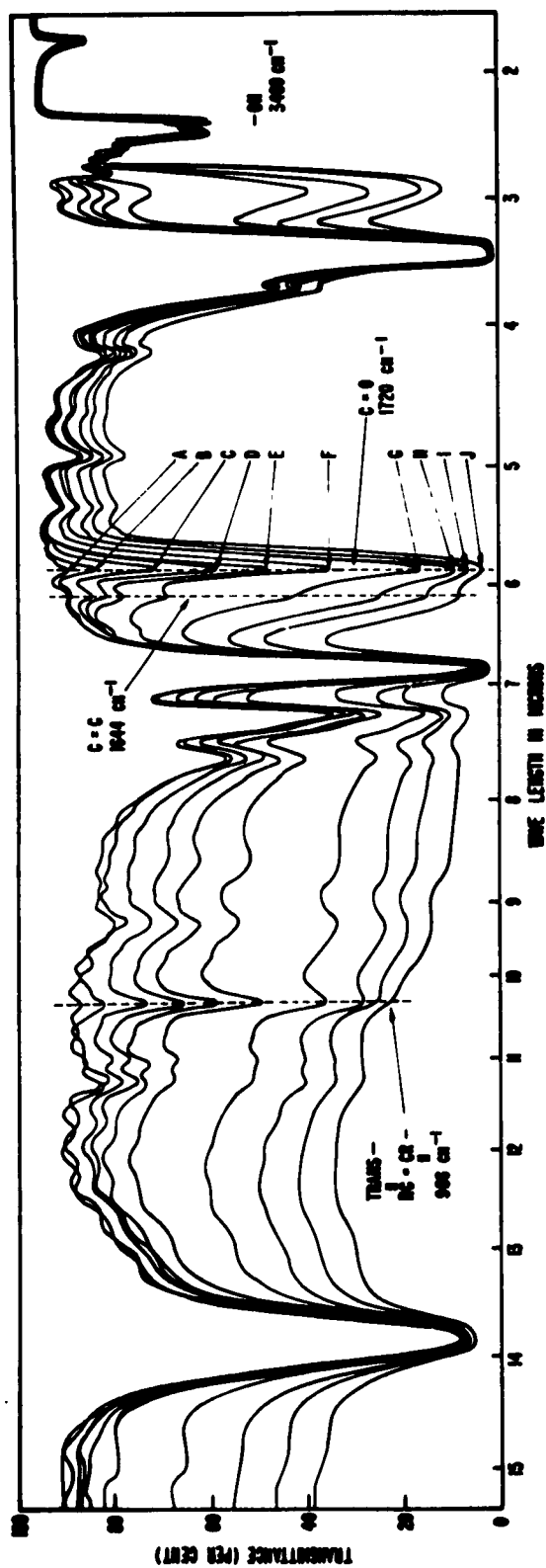


FIGURE 2 INFRARED ABSORPTION SPECTRA OF NON-IRRADIATED VS. ALPHA IRRADIATED POLYETHYLENE

NON-IRRADIATED		ALPHA IRRADIATED	
A	0.32 x 10 ³ ERGS/CM ²	F	0.32 x 10 ³ ERGS/CM ²
B	2.97 x 10 ³ ERGS/CM ²	G	0.36 x 10 ³ ERGS/CM ²
C	0.70 x 10 ³ ERGS/CM ²	H	3.10 x 10 ³ ERGS/CM ²
D	1.02 x 10 ³ ERGS/CM ²	I	7.32 x 10 ³ ERGS/CM ²
E	2.16 x 10 ³ ERGS/CM ²	J	1.40 x 10 ³ ERGS/CM ²

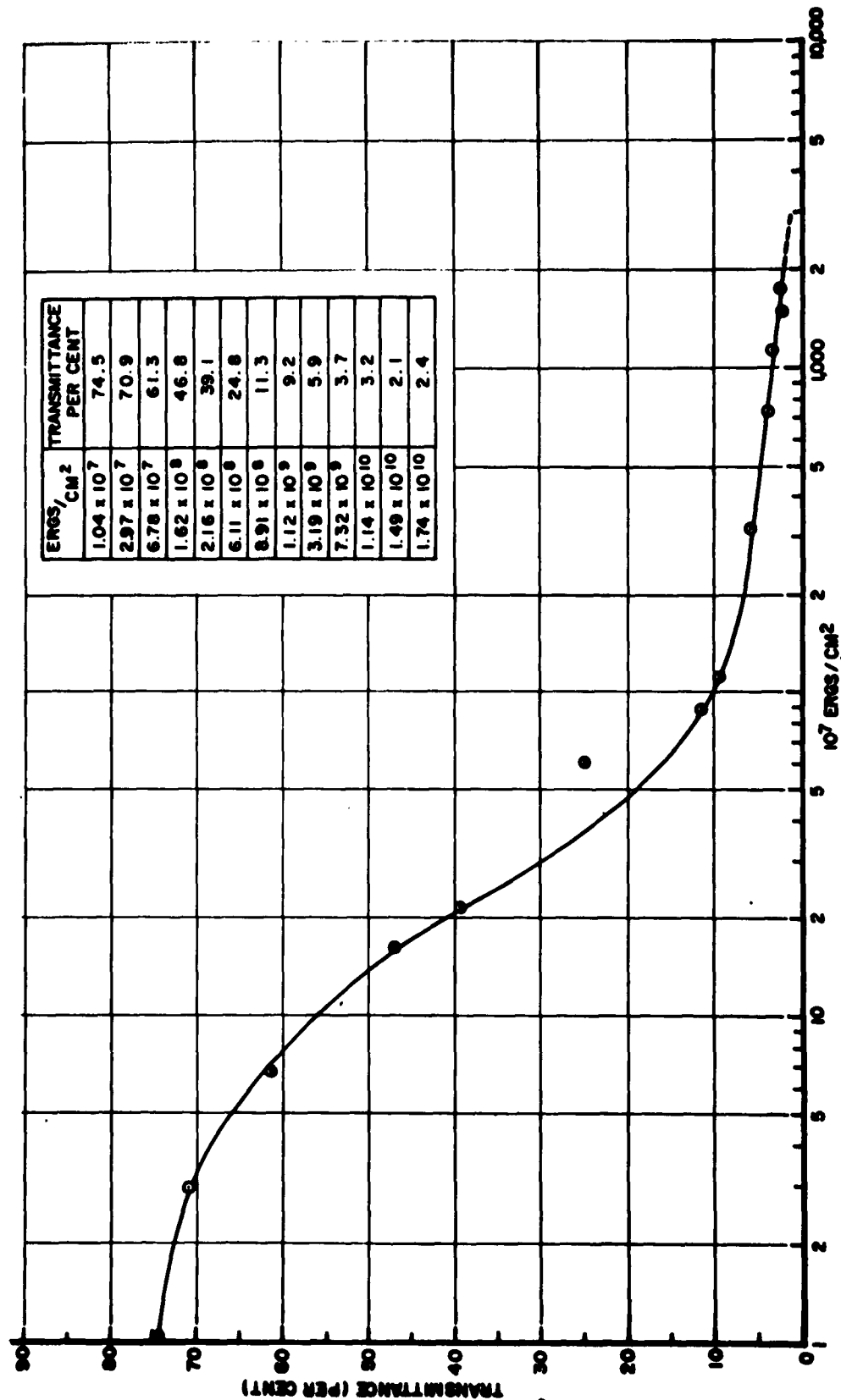


FIG. 3-THE DECREASE IN TRANSMITTANCE AT 1720 CM⁻¹ ATTRIBUTED TO C=O FORMATION IN LOW DENSITY POLYETHYLENE RESULTING FROM ALPHA EXPOSURE.

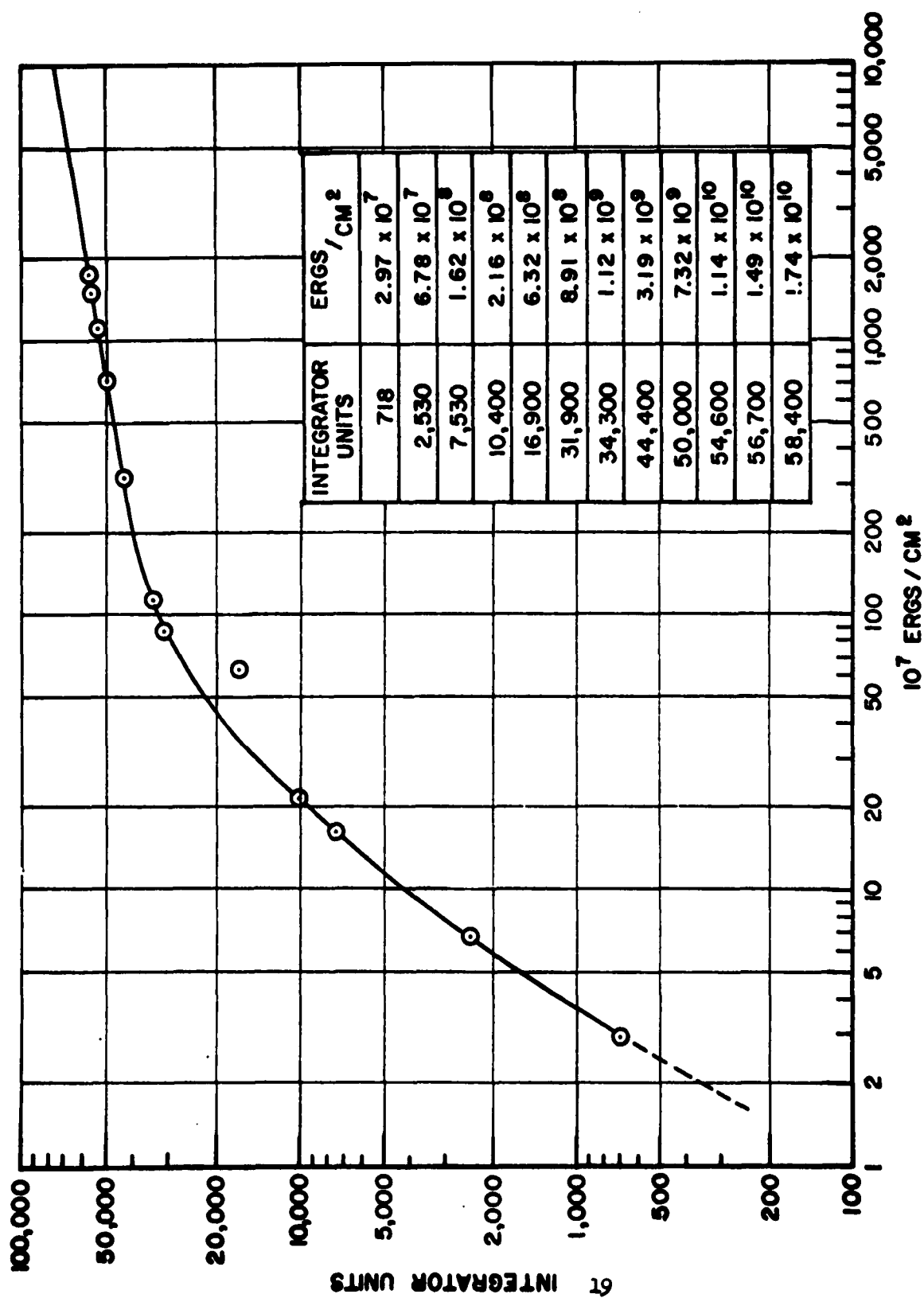


FIGURE 4. THE CHANGE IN THE SPECTRAL AREA OF ALPHA IRRADIATED
VS. NON-IRRADIATED POLYETHYLENE IN INTEGRATED UNITS

component to the infrared spectrophotometer and does increase the time of analysis. These two minor inconveniences are negligible compared to the added advantage of increased sensitivity which the integrator method offers.

Next, in order to measure the chemical changes in the infrared spectrum of polyethylene that are a result of alpha irradiation, differential analysis was performed using the same sample handling techniques and a matched standard sample in the I_0 beam. The samples shown in figure 2 were rerun by this method and are shown in the composite in figure 5. Thus, for the same samples, a larger change in area is obtained for this method as compared to the former. An additional comparison is made in figure 6 utilizing both integrator methods in a plot of the integrator units vs. energy. Thus, one observes that the differential analysis method yields a greater change in the integrator units per unit of exposure energy than the previous method. This method has two distinct advantages. First, the sensitivity of the infrared analysis of the alpha induced changes in polyethylene is increased. Secondly, certain changes in the infrared spectrum, attributed to the effect of alpha particles, are obscured in conventional infrared analysis, but are resolved in the differential spectrum of low density polyethylene. These bands furnish information concerning chemical reactions in the polymer which aid in studying the total alpha induced changes.

Previous gamma exposure studies⁽¹⁾ compared to these alpha exposures on low density polyethylene yield similar absorption characteristics as seen in figure 7. In the so called "finger print" region of the infrared spectra, when comparing alpha vs. gamma exposed polyethylene, one observes what appears to be differences in absorption for the unsaturation bands at 888, 909 and 965 cm^{-1} . These are complex regions to study because of the general increase in absorbance from 800 to 1400 cm^{-1} . The hydroxyl and carbonyl bands for both the alpha and gamma irradiated samples exhibit little difference from each other. However, at 1644 cm^{-1} , there is a C=C absorption in the alpha irradiated samples, which is not significant in the gamma irradiated polymer. One observes C=O absorption at 1720 cm^{-1} in both the alpha and gamma exposed samples, but significant C=C absorption at 1644 cm^{-1} is apparent only in the alpha exposed polymer. Thus, a potential technique exists for distinguishing and monitoring two different types of radiation with one detector material. This

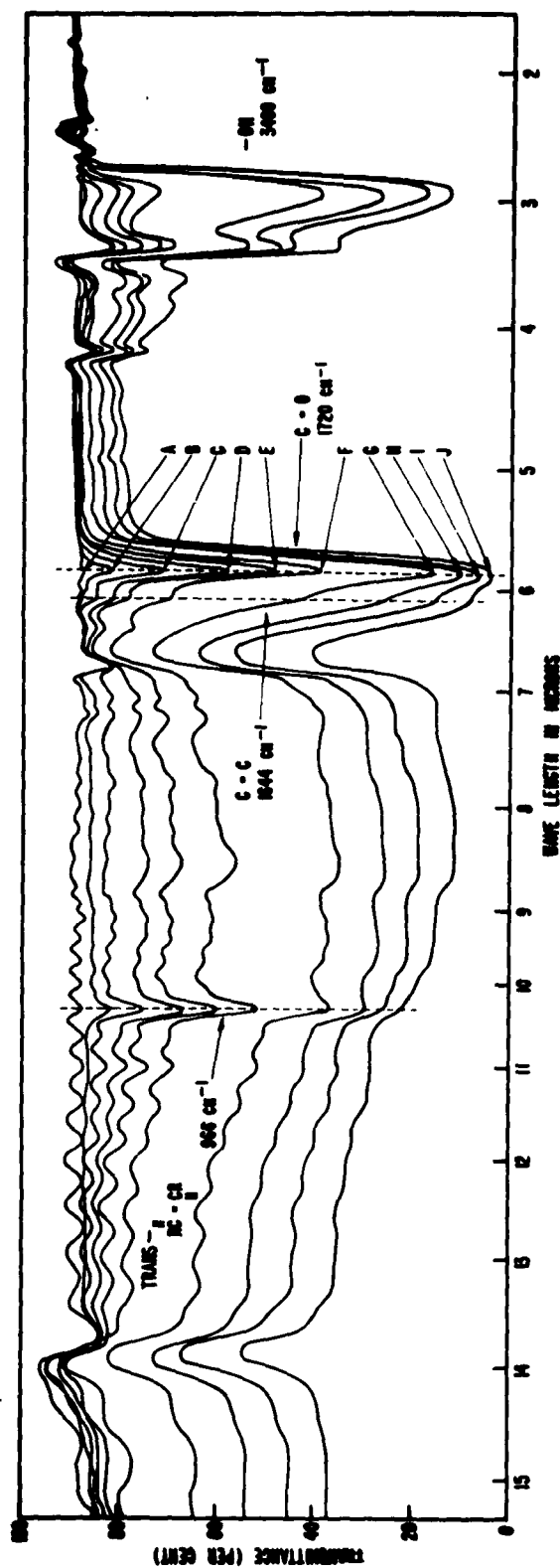


FIGURE 5 - DIFFERENTIAL INFRARED RECORDING OF NON-IRRADIATED VS. ALPHA IRRADIATED POLYETHYLENE

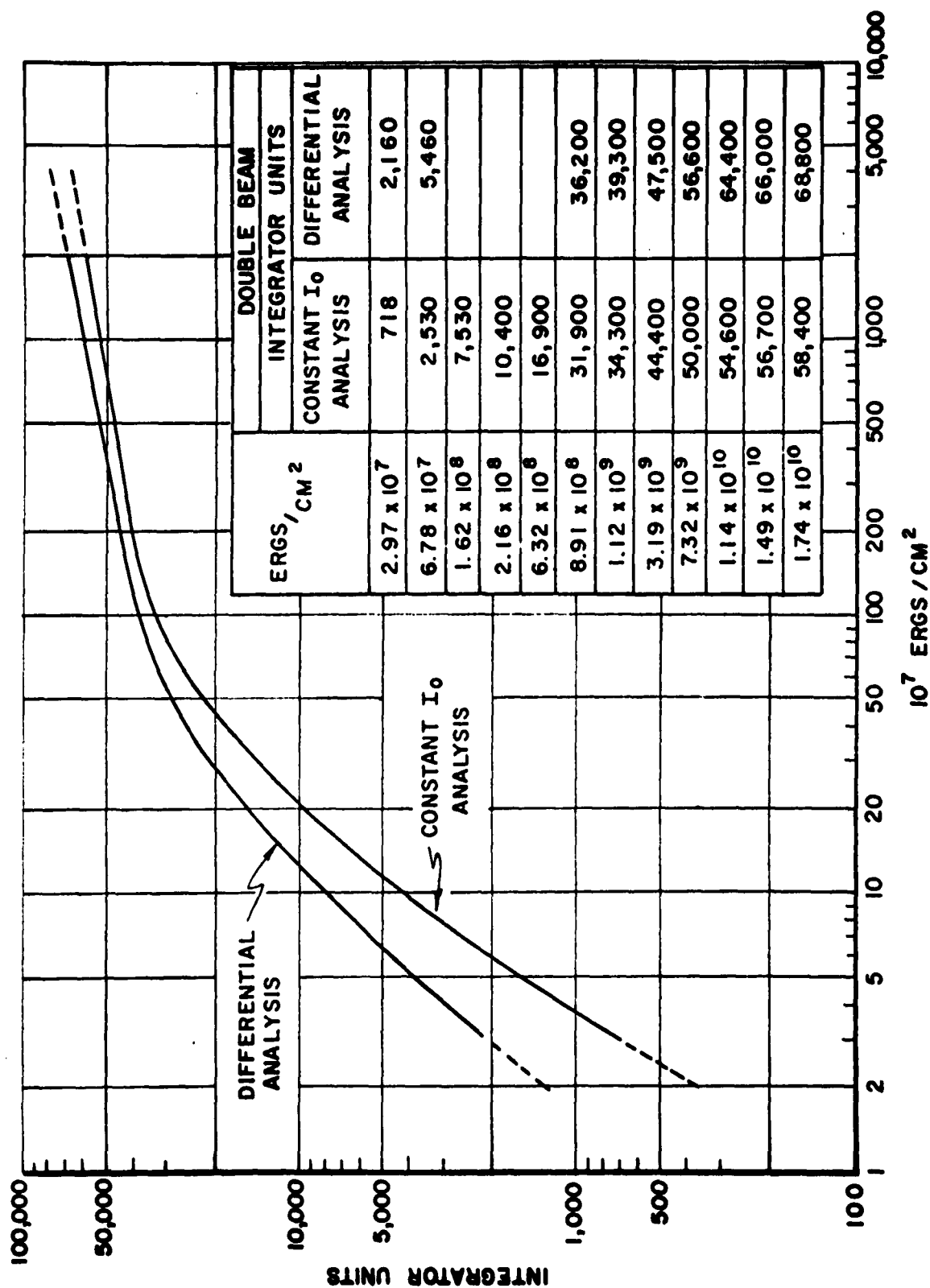


FIG. 6-A COMPARISON OF THE INTEGRATED AREA OBTAINED BY CONSTANT I₀ AND DIFFERENTIAL INFRARED ANALYSIS vs. ALPHA EXPOSURE OF POLYETHYLENE.

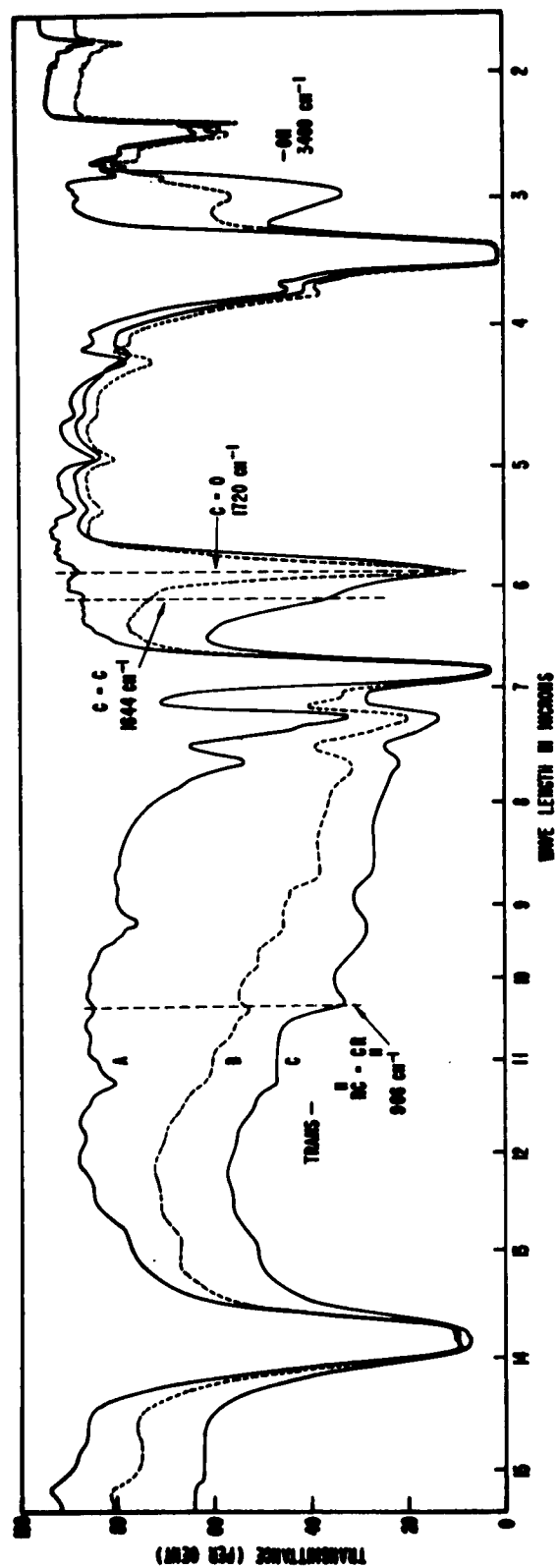


FIGURE 7 A COMPARISON OF THE INFRARED SPECTRA OF ALPHA AND GAMMA IRRADIATED LOW DENSITY POLYETHYLENE

- A. NON-IRRADIATED POLYETHYLENE
 B. GAMMA IRRADIATED POLYETHYLENE $\text{Gy} = 137$ 3.06×10^7 MEV/GM²
 C. ALPHA IRRADIATED POLYETHYLENE $\text{R} = 20$ 1.12×10^6 ERGS/GM²

method utilizes non-destructive analysis and the detector could be used for cumulative exposures. Figure 8 shows the increase in absorption at 1644 cm^{-1} for the alpha irradiated polyethylene as a function of exposed energy.

Thus, it is suggested that one may analyze for the two types of radiation, using a single detector. This may be performed by comparing the 1644 cm^{-1} (C=C) band for alpha energy, to the amount of alpha induced oxidation which would be indicated at 1720 cm^{-1} (C=O). One could then subtract this carbonyl absorbance from the total carbonyl absorbance at 1720 cm^{-1} in the alpha and gamma irradiated sample. The resulting difference will be due to gamma induced oxidation and can be related to exposure dose for gamma rays. Another method would be to place an alpha opaque filter over a portion of the polyethylene before irradiation. After exposure, analyze the protected polyethylene for gamma effects and the open portion for both types of radiation, then relate the difference in alpha energy.

CONCLUSIONS

The report describes the chemical changes detected in the infrared spectra of low density polyethylene, exposed in air to varying doses of alpha particles from polonium-210. Chemical changes induced in this polymer by alpha and gamma radiation are not identical. Thus, a potential method exists for detecting and recording the accumulation of two different types of radiation with a single polymer detector. Alpha particle exposure can be correlated by infrared spectroscopy techniques by utilizing the carbonyl band (1720 cm^{-1}) or the integrated area of the infrared spectra from 666 to 5000 cm^{-1} . Both methods of correlation of alpha exposure to chemical changes are useful. The sensitivity of the infrared analysis can be increased by differential analytical techniques. Thus, the chemical changes induced in polyethylene by alpha particles can be correlated with exposure and can be used for the detection, measuring and cumulative recording of alpha exposures. Given the necessary parameters, the polymer may be used as a secondary calibration technique for polonium-210 alpha sources.

Studies will be made to determine frequency shifts and bands specific to a given type of radiation and to investigate the feasibility of developing a "multi-purpose" polymer for detecting various types of radiation. This polymer detector would indicate total exposed dose as well as that dose attributable to alpha, gamma and neutron radiation, and would be useful for cumulative exposures.

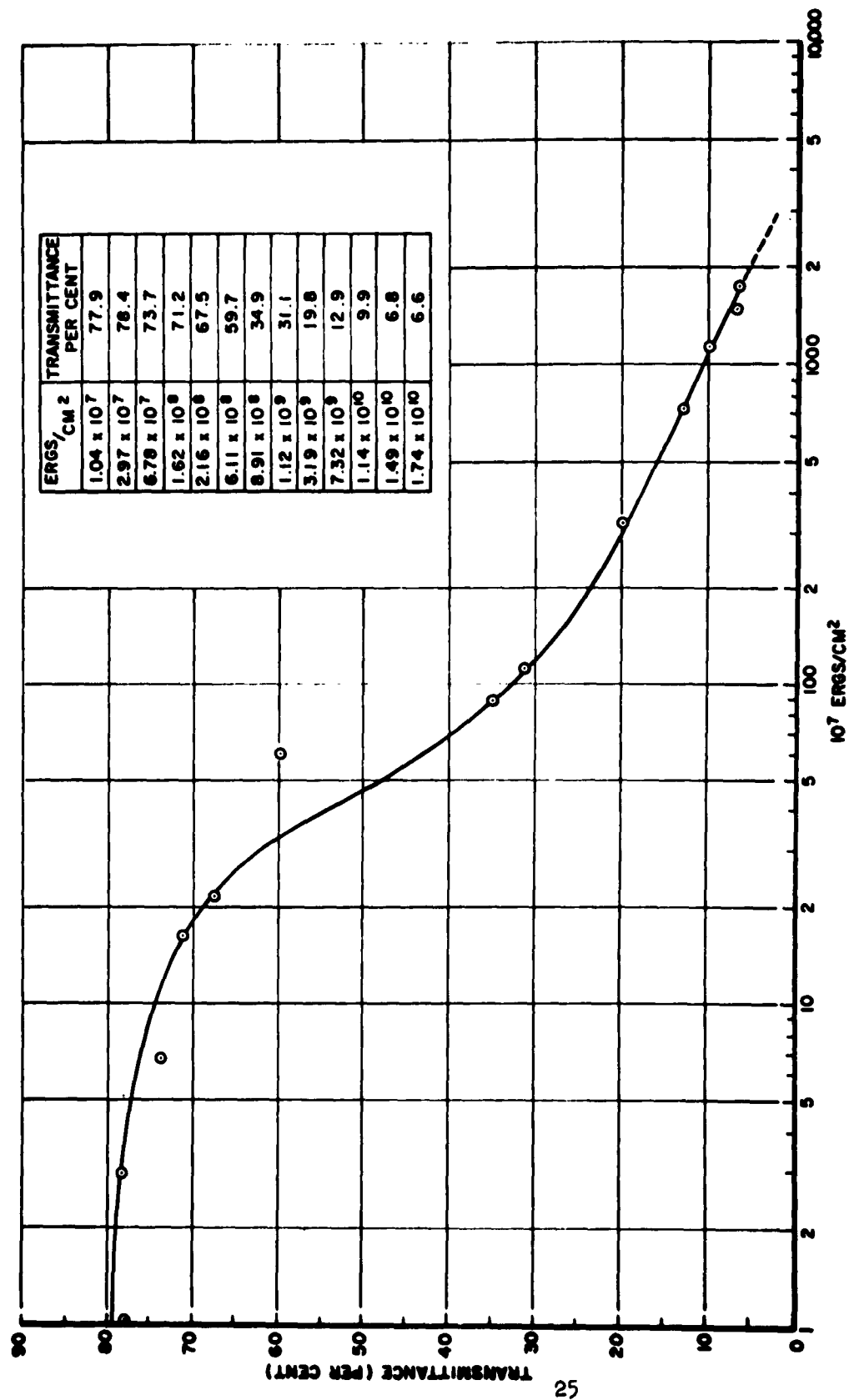


FIG. 8 - THE DECREASE IN TRANSMITTANCE AT 1644 CM^{-1} ATTRIBUTED TO C=C FORMATION IN LOW DENSITY POLYETHYLENE RESULTING FROM ALPHA EXPOSURE.

Further studies on the effect of radiation on low density polyethylene, with incorporated sensitizing materials, will be performed to study the mechanism of the radiation damage.

ACKNOWLEDGEMENTS

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JOE A. SWISHER

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DA Project No. 512-10-001			DA Project No. 512-10-001		
UNCLASSIFIED Report			UNCLASSIFIED Report		
<p>Polyethylene samples were exposed to two different size polonium-210 alpha sources for increasing durations of time. The resulting chemical changes were studied as a function of absorbed energy by means of infrared spectroscopy. The radiation induced chemical changes observed in the infrared spectra of the polymer resulted in an increase in absorption at the characteristic frequencies of the new chemical bonds. Two methods were used to correlate the analytical data with the alpha particle exposure. One method involves the characteristic infrared band frequencies and the other, a unique approach, the correlation of the integrated areas of the infrared spectra. The results indicate the potentiality of polyethylene as a detector for alpha particles and a calibration tool for polonium alpha sources in the range of one millicurie to one curie.</p>			<p>Polyethylene samples were exposed to two different size polonium-210 alpha sources for increasing durations of time. The resulting chemical changes were studied as a function of absorbed energy by means of infrared spectroscopy. The radiation induced chemical changes observed in the infrared spectra of the polymer resulted in an increase in absorption at the characteristic frequencies of the new chemical bonds. Two methods were used to correlate the analytical data with the alpha particle exposure. One method involves the characteristic infrared band frequencies and the other, a unique approach, the correlation of the integrated areas of the infrared spectra. The results indicate the potentiality of polyethylene as a detector for alpha particles and a calibration tool for polonium alpha sources in the range of one millicurie to one curie.</p>		
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